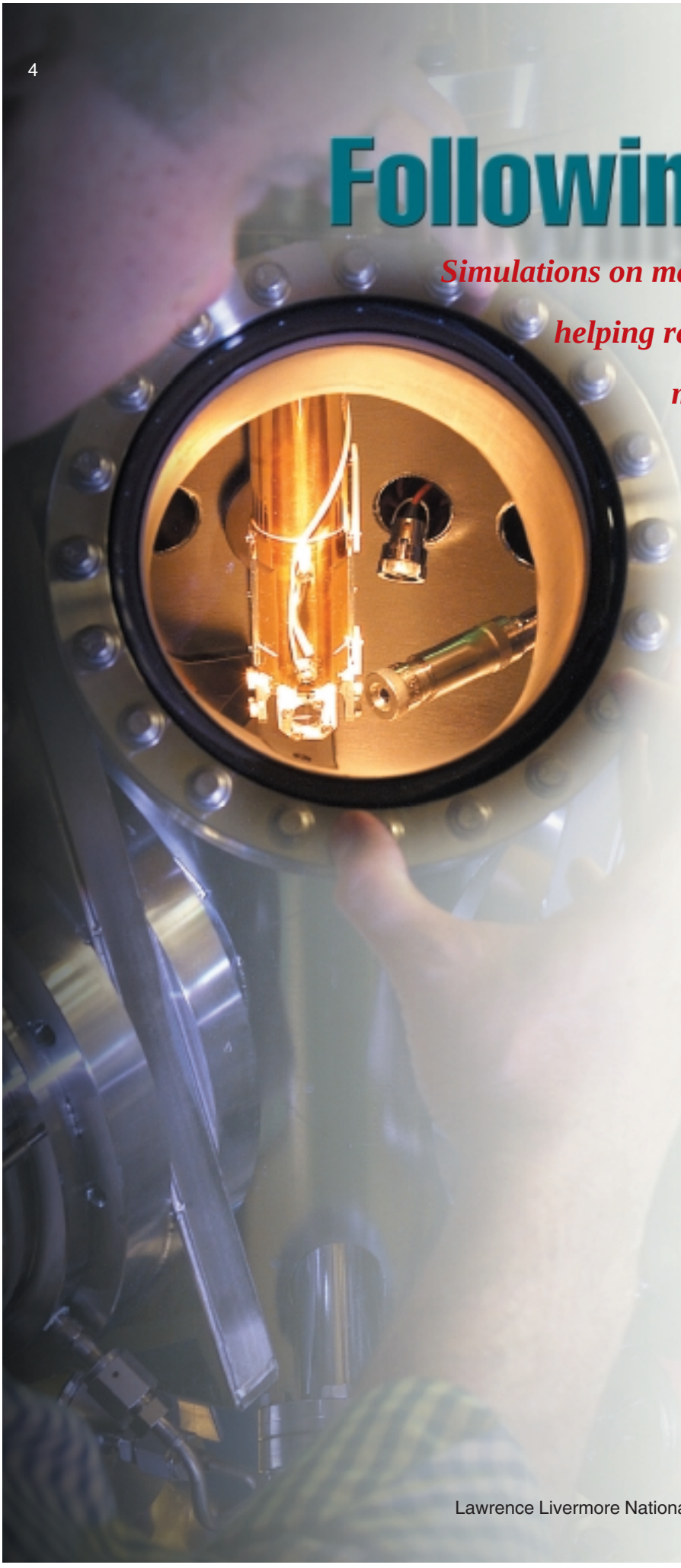
Following Materials

Simulations on massively parallel supercomputers are helping researchers understand how and why materials change and ultimately fail.



FROM breakthrough computer chips to new alloys for automobile engines, advances in materials shape our day-to-day lives and drive economic growth. For Lawrence Livermore scientists, predicting the performance of advanced materials is essential to success in countless research programs in national security, lasers, energy, and biotechnology.

In the past, advances in materials were accomplished by extensive laboratory testing combined with a healthy dose of guesswork, a time-consuming and often costly approach. Recently, a team of Lawrence Livermore scientists has begun to tap the vast computational power of Department of Energy supercomputers to simulate and accurately predict the performance of advanced materials. These simulations cover a wide range of operating environments and length and time scales.

The new field, called computational materials science, is one of the fastest growing areas within the field of chemistry and materials science. Leading the Livermore effort is materials physicist Tomas Diaz de la Rubia. He heads a team of 20 materials scientists, metallurgists, chemists, computer scientists, and physicists as well as graduate students from the United States and Europe.

The simulation research team works closely with Livermore computer scientists and collaborators from U.S. universities such as Princeton, Washington State, the University of Pennsylvania, MIT, and UCLA, and American companies such as IBM, Intel, and Applied Materials. The work is funded by Lawrence Livermore research programs, the Laboratory Directed Research and Development Program, the Department of Energy's Office of Science, and U.S. industry.

Diaz de la Rubia notes that Livermore research programs need many different classes of materials—including ceramics, glasses, plastics, and metals—and biological products. As a result, his group has assembled a broad research portfolio, one that includes simulating the mechanical properties of materials under extreme conditions, the accumulation of radiation damage in metals, the interaction of laser light with glass, the integrity of canisters for nuclear waste

over Time and Space

storage, the design and properties of advanced alloys, the actions of biomaterials such as enzymes, the dynamics of metal corrosion and cracking, and the diffusion of dopants in semiconductor manufacturing.

A Major International Presence

While helping Lawrence Livermore programs, the group is also establishing a major presence in the scientific community. The team publishes frequently in scientific journals and is prominent at international conferences. For example, Diaz de la Rubia is co-chair of the spring 2001 meeting of the Materials Research Society, which is expected to attract more than 3,000 scientists from around the world to San Francisco.

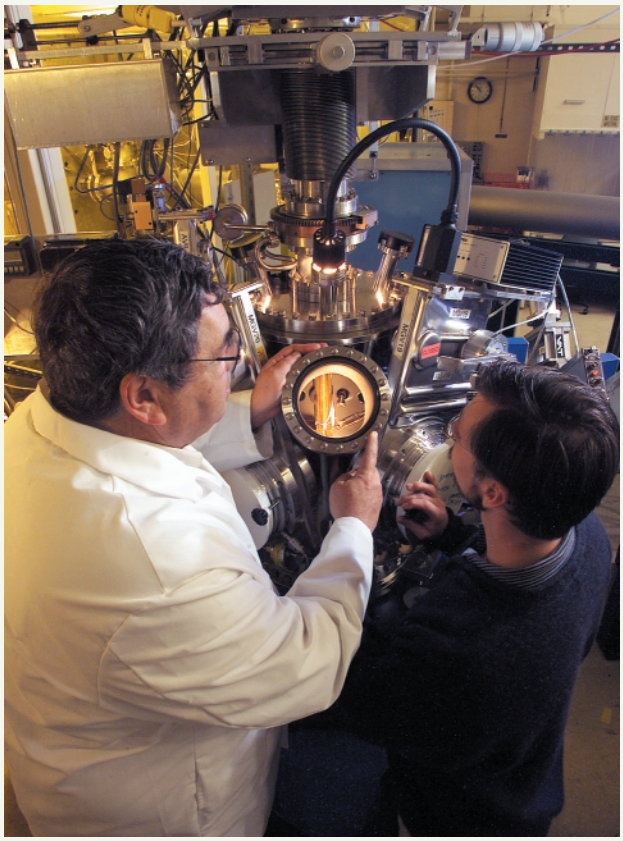
The Livermore work is part of DOE's Office of Science Computational Materials Science Network, composed of multidisciplinary scientific teams at several national laboratories. The network fosters basic materials science research within DOE and also serves to attract the best new talent to the discipline.

Diaz de la Rubia notes that scientists have long used computers to model materials and their performance. However, the newest generation of supercomputers, which employ thousands of microprocessors to tackle simulations once considered intractable, represents a significant advance. The computational materials science team works closely with members of the Laboratory's Center for Applied Scientific Computing to develop simulation tools that take advantage of multiprocessor supercomputers. The most advanced of those computers were acquired through DOE's Accelerated Strategic Computing Initiative (ASCI), a key component of the Stockpile Stewardship Program to assure the safety and performance of the nation's aging nuclear stockpile.

Writing or adapting codes for large parallel machines, in which complex problems are divided up to be jointly solved by multiprocessors, is a large effort. "The task requires people to think about software in new ways," says Diaz de la Rubia. The payoff, however, is a significant improvement in performance, as measured in the time required to run a model and the number of atoms being simulated. On Livermore's ASCI Blue supercomputer, about 350 million atoms could be simulated, but the newest computing platform, ASCI White, will track 10 billion atoms simultaneously.

The Livermore simulations are closely linked to laboratory experiments on the same materials. Experimental data ensure that the simulations accurately reflect the materials' chemical, mechanical, and thermodynamic properties as well as their manufacturing methods and the ways by which they age and ultimately fail. Only with solid experimental underpinnings, says Diaz de la Rubia, can the models help advance the understanding of how materials form, how they react under changing conditions (especially extreme environments), and how they can be improved.

The codes are so sophisticated that Livermore researchers are beginning to predict what scientists will see when imaging materials through electron microscopes. Their simulations constitute an important bridge between



Livermore materials simulations are closely coupled to a program of laboratory experiments. Researchers Mike Fluss (left) and Brian Wirth measure the atomic transport properties of radiation damage defects in metals, including plutonium; the data are used to refine codes that simulate and predict the performance of stockpiled nuclear weapons.

computer models and experiments. When a computer-generated image of calculated defects in a crystal is superimposed on an image of the same material taken with a transmission electron microscope, the results are remarkably similar. "When we see a certain image in the microscope, we now have a good idea about the mechanisms that produced it," says Diaz de la Rubia.

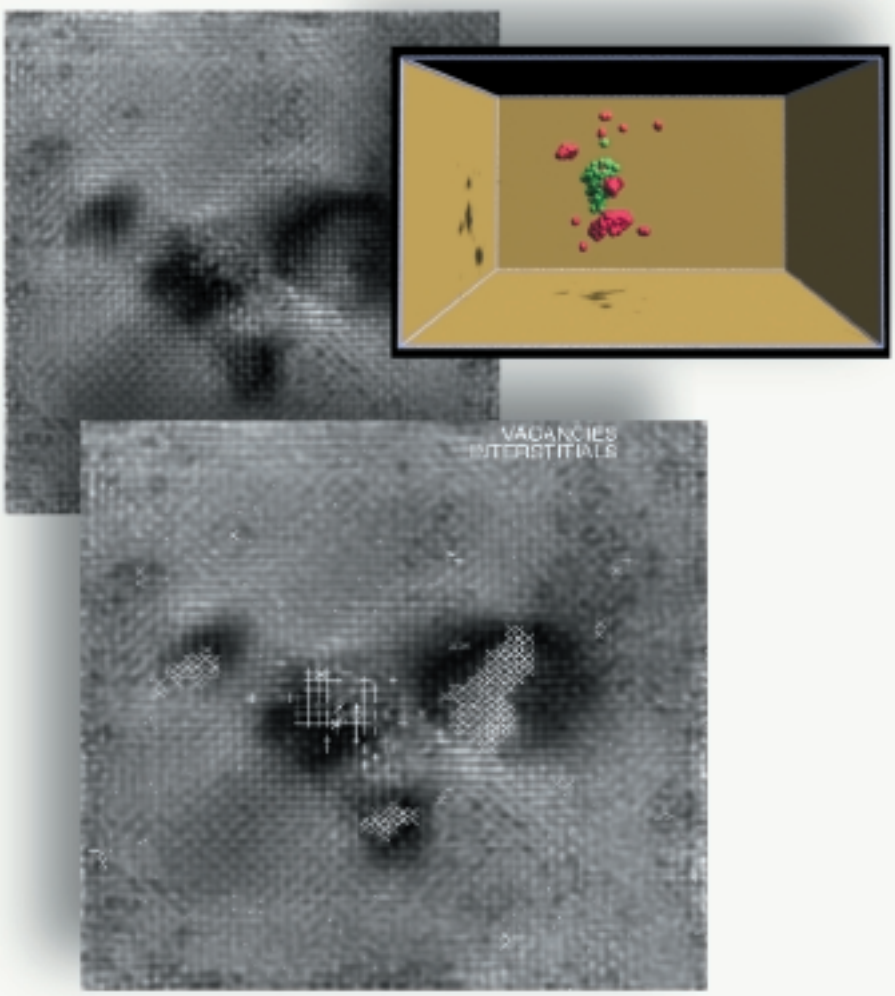
Multiscale Modeling of Materials

The Livermore codes incorporate multiscale modeling, an approach that is needed because material properties often depend on phenomena that take place at all length scales, from nanometers to meters. Multiscale modeling also incorporates a range of time scales from billionths of a second to tens of years. By combining models that cover the full range of length and time scales, scientists can simulate the evolution of mechanical and chemical changes in materials. Such changes may start with a defect occurring in a metal's crystalline lattice over a few nanometers in length and in a billionth of a second. And yet, multiplied millions of times over several years, such defects may combine to cause catastrophic failure of a computer chip, a glass optic, or a pressure vessel.

The most general approach to multiscale modeling is called information passing. In this method, which was introduced to the Laboratory by materials scientists Wayne King and David Lassila, simulations of matter at one scale are based on the results of simulations at a lower (more finely detailed) scale. The challenge is to form a coherent simulation of a material by building what Diaz de la Rubia calls "hooks between different scales."

The other approach, applicable in a limited number of cases, is called embedded multiscale modeling; its development at Livermore is being spearheaded by physicists Bernie Alder, Robert Rudd, Andrew Quong, and Vasily Bulatov. It uses the same code to solve the motion of molecules at all scales, with different physical models having different levels of approximation at each scale. For example, the way a nanometer-size crack behaves at the smallest lengths is similar to the way larger-scale cracks behave. Thus, it is a candidate for embedded multiscale modeling.

There are four major length scales: atomistic (measured in nanometers), microscale (micrometers), mesoscale (hundreds of micrometers) and



The Livermore simulations are beginning to predict the images that scientists will see when viewing materials through electron microscopes. On the upper left is a high-resolution transmission electron micrograph of a crystal of gold showing clusters of defects, namely interstitial atoms (atoms out of place, in red) and vacancies (points in the crystalline lattice where no atoms reside, in green). At the upper right is a computer simulation of how the defects should appear through the electron microscope. Superimposing the simulated image onto the electron micrograph (lower center) shows an impressive correlation between the two.

continuum (larger than 100 micrometers). In most cases, all four scales must be used to completely model a material's performance over a selected period of time. Each scale also requires a special code. For example, the microscale uses new codes such as micro3D, developed at Lawrence Livermore in collaboration with Washington State University researchers, while the continuum scale uses ALE3D, also developed at Livermore.

Simulations Begin with the Atom

The atomistic scale, which involves the properties and interactions of electrons and atomic nuclei, has been studied in great detail since the discovery of quantum mechanics. Atomistic simulations based on so-called first principles help scientists understand how atoms are arranged in crystals, how they bond to other atoms, and how impurities affect them. It is impossible, however, to use only atomistic models to simulate all material properties because of the sheer number of atoms that would be required for such a simulation.

Microscale simulations determine many materials properties, such as strength. But until the advent of ASCI machines, computers did not have the computational horsepower to perform microscale simulations with great fidelity. "People had been studying the collective behavior of dislocations in crystals for 40 years, but they couldn't simulate it until now," says Diaz de la Rubia. The microscale, he says, is the critical link between the atomistic and mesoscopic scales.

Likewise, the mesoscale links the microscale to the continuum scale. The mesoscale determines the structure of grains (a portion of a crystal in which all the atoms are oriented the same). Computer codes essential to understanding the performance of many nuclear weapons are based on this scale.

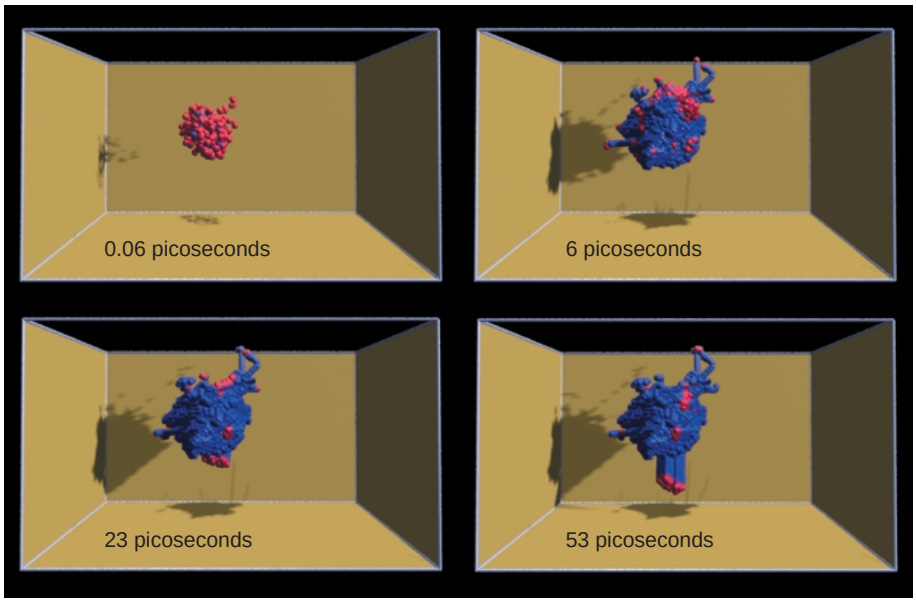
Finally, simulations on the continuum scale provide engineers with the likely behavior of materials in such areas as the damage that occurs in crash testing. At this level—visible to the naked eye—simulations using finite-element methods and experimentally derived equations model physical structures with element sizes that approach those studied individually with mesoscale models.

One essential aspect of multiscale modeling is its interdisciplinary nature. At Livermore, the effort is led by physicist Elaine Chandler and brings together scientists in the Defense and Nuclear Technologies, Physics and Advanced Technologies, Engineering, and Chemistry and Materials Science directorates to accomplish the goals of the modeling programs.

A major focus of the collaborative multiscale modeling effort is predicting the strength of metals in a variety of environments. Diaz de la Rubia notes that a general relationship between a metal's composition and its strength has been well established for decades. However, a detailed understanding of the mechanisms that confer strength based on the metal's crystalline microstructure has been lacking. Such knowledge is particularly vital to making predictions of performance under extreme conditions of pressure and temperature. "We want to predict how metals will deform under all conditions, and multiscale modeling is showing us the way," Diaz de la Rubia says.

Dislocations Are Key

He explains that in a metal, atoms are stacked in an ordered, crystalline lattice. However, there are also regions of less ordered stacking that can affect mechanical properties. These disordered areas can be caused by impurities, point defects in the crystal's lattice, or especially dislocations, which are misaligned planes of atoms that are sometimes referred to as line defects. When dislocations move through a crystalline lattice, they create plastic deformation, the ability of a material to bend without breaking. Without some plasticity, a metal becomes brittle and vulnerable to fracture and failure.



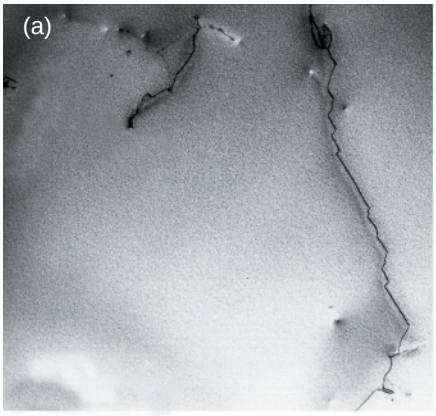
Atomistic simulations of radiation damage in copper atoms show the accumulations of defects over time, particularly those caused by clusters of interstitials (atoms that are not in their correct place in the copper crystal) or by replacements of the original atoms in the lattice. Data from atomistic simulations are used as the basis for microscale simulations.

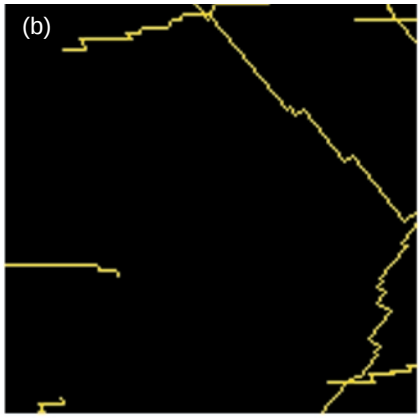
Livermore microscale simulations model the motion, multiplication, and interaction of dislocations, a phenomenon called dislocation dynamics. The simulations show the collective and complex behavior of millions of dislocations per cubic centimeter and how they determine a material's plastic deformation. The simulations are helping researchers better understand how certain manufacturing methods, such as the addition of selected impurities (alloys), harden a metal by interfering with dislocation movement.

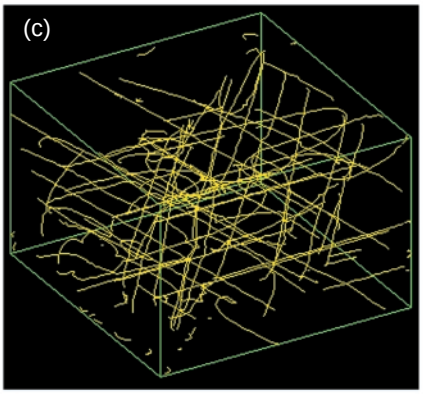
The strength simulation work is closely tied to experiments. The overall effort is headed by David Lassila and is based on the behavior of dislocations in molybdenum and tantalum crystals because these crystals are similar to materials found in many Laboratory programs and the nuclear stockpile. The experiments provide well-characterized dislocation structures to validate the results of the simulations. The experimental results are aided by directly observing dislocation mobility with transmission electron microscopes.

The current parallel version of the simulation code has been run on Livermore's ASCI Blue supercomputer using 80 processors and 260 hours of processing time. Led by Lassila and Bulatov, the researchers are revising the code to run on ASCI White. Lassila and Bulatov, together with physicist John Moriarty, are looking to extend the simulation work to areas of extreme pressure and temperature to improve the accuracy of current material models.

Predicting the strength of materials is important in almost all aspects of stockpile stewardship. Indeed, stockpile stewardship–related projects comprise the majority of the group's work. Diaz de la Rubia notes that materials are at the heart of most of the issues associated with the nation's enduring nuclear stockpile and that a central goal of ASCI is to acquire full-scale materials simulation tools to more accurately predict the lifetimes of weapon components. In particular, scientists want to improve their ability to predict the effects of aging or the performance of a remanufactured weapon part (see *S&TR*, June 1999, pp. 22–25).







(a) An electron micrograph of molybdenum under pressure determines (b) the initial conditions used in the model for simulating dislocation dynamics. (c) The simulation shows a compressed sample of molybdenum with many dislocations running through the crystalline lattice.

Radiation Damage Is Multiscale

One of the most challenging stockpile stewardship–related simulation problems is radiation damage to warhead components and materials. The problem is inherently a multiscale phenomenon. The individual events occur over a scale of 100 nanometers and a small fraction of a second, but the effects build up over decades throughout a material (see box on p. 10).

The materials scientists are applying multiscale modeling to predicting the performance of nonweapons materials in nuclear radiation environments. One team led by physicist Maria Jose Caturla and materials scientist Brian Wirth is modeling materials that could be used in future fusion reactors. Another team headed by physicist Patrice Turchi is modeling the long-term performance of waste canisters manufactured from exotic metal alloys. The canisters are being considered for storage of high-level nuclear waste at Yucca Mountain, Nevada (see *S&TR*, March 2000, pp. 13–20). Livermore scientists are performing simulations to determine how the canister will react over a 100,000-year period in response to extreme conditions of high humidity, temperature, and radiation. The assignment is a huge challenge because

it involves length scales ranging from the atomistic to the entire 45,000-kilogram, 3-meter-long waste canister.

One related area of interest is stress corrosion cracking, the most common mode of failure for a wide range of materials, including not only nuclear waste containers, but also bridges, fiber-optic cables, and nuclear reactor pressure vessels. Because microscopic cracks grow slowly, it is difficult to predict when a part will fail. For the first time, Livermore scientists led by Andrew Quong and materials scientist Wayne King are making the connection between the breaking of single atomic bonds at the tip of a growing crack and the ultimate failure of a part. They are planning to further their research through collaborations with materials scientists from IBM's Almaden Research Center in San Jose, California.

Advancing Semiconductors

An area of great potential is applying multiscale modeling to gain a better understanding of processes in the semiconductor industry. Diaz de la Rubia explains that the semiconductor industry has traditionally used simple models to explain the actions of dopants (impurities deliberately added to semiconductors to achieve a desired property) and defects. However, semiconductor manufacturing involves more than 200 steps in a process that is far too complex to be treated with these models. What's more, the industry is building increasingly smaller features on their chips, and the lack of accurate modeling, particularly on the atomistic scale, is proving to be a serious impediment to the development of next-generation devices. As a result, new atomistic-level models are required.

"Semiconductor companies want to be able to predict the outcome of a process from a given set of manufacturing conditions," says Diaz de la Rubia. To aid the manufacturers, Livermore researchers investigated the kinetics of defect and dopant migration in silicon chips, using funding from DOE's Office of Science and U.S. corporations such as Intel and Applied Materials. The approach provides a fundamental database for use in developing predictive simulations. It combines both experiments and theoretical advances and uses the same methods applied to other multiscale modeling projects.

The simulation results, when compared to results from laboratory experiments, showed the models to be very accurate. The project's success and the promise of the Livermore approach led to a Cooperative Research and Development Agreement with Intel and Applied Materials to create predictive modeling tools for semiconductor manufacturing.

In like manner, says Diaz de la Rubia, researchers are creating a family of tools for simulating biochemical processes. These tools will permit more accurate and rapid protein structure predictions. They will contribute to the study of DNA and aid predictions of protein function, drug activity, and the effects of chemical hazards such as carcinogens.

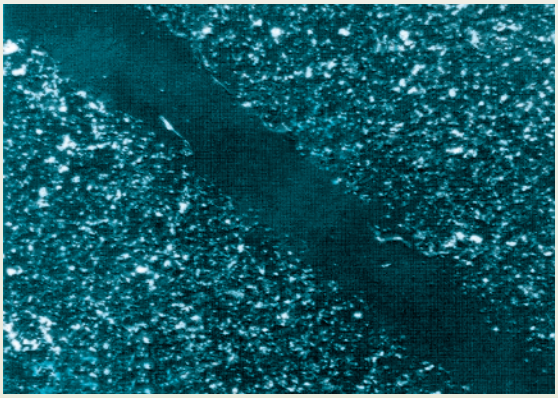
Materials scientist David Lassila, left, discusses the results of dislocation dynamics simulations with his team while they are directly observing dislocations with a transmission electron microscope. Mark Wall is seated at the transmission electron microscope, while Mary Le Blanc and R. Ann Bliss look on at right.

Simulations Reveal Damage from Radiation

Radiation damage is a ubiquitous phenomenon that affects many Lawrence Livermore research programs, the nation's nuclear weapons stockpile, and the nuclear power industry. Radiation damage can significantly degrade mechanical properties, seen most notably in increased brittleness and outright failure of a component.

The damage shortens the lifetime of pressure vessels in nuclear power plants and limits the choice of materials for fusion energy research. The problem is of particular concern for weapons materials such as plutonium. When plutonium-239 decays, it emits an alpha particle (a helium nucleus) and an atom of uranium-235. The resulting buildup of gaseous helium atoms and displaced plutonium atoms from the recoiling uranium could produce unacceptable changes in the plutonium metal. (See *S&TR*, June 2000, pp. 15–22.)

Although radiation damage has been studied for many years, the underlying mechanisms that relate damage to degradation of mechanical properties had not been clearly demonstrated until recently by a Livermore team. Headed by Diaz de la Rubia, the team reported its findings in the August 24, 2000, issue of *Nature* magazine. Because the damage evolves over a wide range of length and time scales, Diaz de la Rubia believed it could only be understood through multiscale modeling whose results are then validated by laboratory experiments.

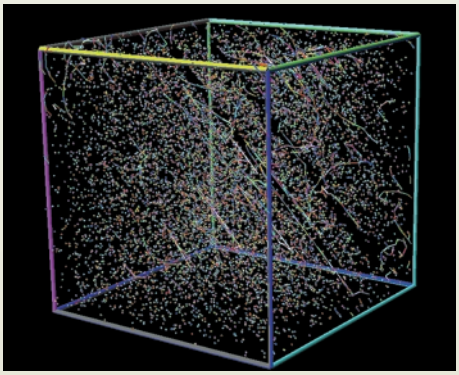


Simulations show that some of the dislocations in irradiated copper form defect-free channels.

The team used atomistic and then three-dimensional microscale simulations of irradiated metals. The simulations showed defects (both vacancies and out-of-place atoms) accumulating at the shortest scales (nanometers and picoseconds) and quickly growing into clusters. These clusters served to pin or prevent the movement of dislocations, which are misaligned planes of atoms. The free movement of dislocations confers plasticity or strength properties on metals.

The simulations showed that as some of the dislocations moved through the metal, they annihilated any defects along their path, thereby forming 200- to 300-nanometer-wide channels along particular directions (see figure below, left). The defect-free channels resulted in plastic instability, that is, areas of reduced strength. The team then compared the simulations to images taken with electron micrographs of irradiated copper and saw a very close correlation between the two (see figure below, right). While electron micrographs had previously revealed the presence of clear channels, scientists could not explain their formation.

Diaz de la Rubia notes that in the case of modeling radiation damage in metals, length scales are not as important as the time scales and energy scales of particles (for example, neutrons) involved in causing damage. The critical information, he says, is "figuring out how fast the changes happen so we can make predictions." To accomplish that requires first simulating the evolution of the smallest changes at the atomistic scale, using kinetic Monte Carlo software, and then applying the results as input to dislocation dynamics models to determine how mechanical changes occur.



Electron micrographs of irradiated copper reveal the same clear channels predicted by the simulations. The clear channels are areas of reduced strength.

Another major area of materials simulation work is for the National Ignition Facility (NIF), which will be the world's largest laser and is now under construction at Lawrence Livermore. Because the facility will have the largest concentration of optics anywhere, scientists need a more accurate method for predicting how the extremely pure optical glass will respond to the laser's high fluence levels.

Diaz de la Rubia is embarking on an effort to predict the lifetime of NIF optics with experiments and multiscale modeling. The project focuses on the final optics system, where the frequency of each laser beam is tripled into the ultraviolet with crystals of potassium dihydrogen phosphate and then focused onto a target through a fused silica lens. "We want to be able to model the response of fused silica in the presence of high intensity laser light," he says.

The modeling effort, led by physicist Michael Feit, involves performing atomistic calculations based on the fundamental properties of silica when exposed to laser light and the methods by which cracks propagate in glass. All of the information will be passed up to continuum-level simulations using the ALE3D code to model large-scale fractures. As with other multiscale modeling work, the NIF project involves a multidisciplinary team that includes experts in optics, lasers, materials science, and mechanical engineering.

With the advent of multiprocessor supercomputers, scientists can finally execute simulations with unprecedented accuracy and with seamless integration over all length and time scales. The Livermore simulation studies will help stockpile stewardship scientists to confidently predict the performance of stockpiled weapons.

The multiscale modeling work affects much more than DOE research programs. Use of the supercomputers amounts to a revolution in the materials science community. In this new era, simulation will guide advanced materials development and will show how materials form, how they react under changing conditions, and how they can be optimized for better performance. Simulations will also provide basic information about material behavior of interest to the nation's industrial products manufacturers.

—Arnie Heller

Key Words: Accelerated Strategic Computing Initiative (ASCI), ASCI Blue, ASCI White, Center for Applied Scientific Computing (CASC), dislocation dynamics, Laboratory Directed Research and Development, multiscale modeling, National Ignition Facility (NIF), plutonium, radiation damage, stockpile stewardship, Yucca Mountain.

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About the Scientist

TOMAS DIAZ DE LA RUBIA is the deputy division leader for Science and Technology in the Chemistry and Materials Science Directorate. After completing his Ph.D. in physics at the State University of New York at Albany, he joined Lawrence Livermore in 1989 as a postdoctoral fellow working on supercomputing applications for fusion materials research. On becoming full-time staff, he first developed physics-based predictive process models for semiconductor manufacturing, in collaboration with several large semiconductor corporations. Then he began developing multiscale models of materials strength and aging in irradiation environments. He has published over 100 peer-reviewed scientific articles, chaired numerous international conferences and workshops, edited conference proceedings and special journals, served on national and international panels, and is currently on the editorial board of five scientific journals and a member of the American Physical Society's selection panel for the Rahman prize in computational physics. His research interests are in applying large-scale computing to materials problems, developing predictive tools for materials performance, and designing new methodologies for multiscale modeling.